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TRANSPORT METHODS USED FOR HOMELAND SECURITY AND NON-PROLIFERATION DETECTION APPLICATIONS: AN OVERVIEW

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ABSTRACT

This paper discusses recent investigations in homeland security and non-proliferation research that involve the application, analysis, and experimental validation of transport computations as they are applied to SNM detection scenarios. The use of transport theory methods, particularly 3-D Deterministic radiation transport, as a tool in assessing scenarios involving Special Nuclear Materials (SNM) detection for homeland security and non-proliferation applications has increased in recent years. This is primarily due to the need to optimize detectors and understand radiation signatures to overcome the many challenges associated with SNM detection, and the complex nature of the radiation spectra involving neutron and gamma transport in detection scenarios. Augmenting the effort is the more-ready access and availability of high performance computing. Overall, the valuable information obtained from the application of deterministic and Monte Carlo Transport methods, and how those methods are being used to augment detection capabilities is discussed.

Key Words: 3-D Transport, Computational Models, Non-Proliferation

1. INTRODUCTION

The reliable detection of neutrons (*n*), gamma rays (γ-rays), and alpha particles (α-particles) is of great importance in preventing the proliferation weapons of mass destruction and terrorism. The danger of nuclear materials proliferation and unauthorized shipments of controlled materials cannot be underestimated or overstated. Therefore, simulation of active and passive interrogation techniques for the detection of Special Nuclear Materials (SNM) are being pressed among many applications in the areas of nuclear safeguards, nuclear nonproliferation, and homeland security.[1,2,3] The performance assessment of existing techniques and the development of new, more advanced ones rely on accurate simulation of realistic threat scenarios. The use of transport theory methods, particularly 3-D deterministic radiation transport, as a tool in assessing scenarios involving Special Nuclear Materials (SNM) detection for homeland security and non-proliferation purposes has increased in recent years. This is primarily due to the challenges involved in SNM detection, the more ready access and availability of high performance computing platforms, and attempts to understand the complex nature of the source radiation spectra involving neutron and gamma transport in detection scenarios.[4] This paper is organized as follows: we briefly discusses the principal issues related to detection modalities, transport methods, and cross section considerations; also addressed is total source construction, and forward and adjoint radiation transport methods. The remainder of the paper discusses detection modalities, recent applications, conclusions and references.

2. DETECTION MODALITIES

Detection modalities for SNM detection primarily pertain to the assessment of passive neutron and gamma ray signatures, although active interrogation is slowly becoming more evident as a possible mode. Due to the fact that neutrons have no charge, they cannot be directly detected, and thus neutron detection techniques are based on nuclear reactions that create charged particles which can, in turn, be detected by radiation detectors. Slow neutron detectors, designed for neutron energies below the cadmium cutoff of ~0.5 eV, are detected via nuclear conversion reactions, such as (*n,α*) and (*n,p*) reactions. All common techniques used to detect slow neutrons result in heavy charged particles[5] as shown in Figure 1-1.

$$\text{Target nucleus} + \text{neutron} \rightarrow \begin{cases} \text{recoil nucleus} \\ \text{proton} \\ \alpha\text{ - particles} \\ \text{fission fragments} \end{cases}$$

Figure 1-1. Common reactions used for slow neutrons detection.

The three "conversion reactions" commonly used in neutron detectors are [6]:

$$ {}^{10}_{5}B + {}^{1}_{0}n \rightarrow \begin{cases} {}^{7}_{3}Li + {}^{4}_{2}\alpha & Q = 2.792\ MeV\ \text{(ground state, 6\%)} \\ {}^{7}_{3}Li^{*} + {}^{4}_{2}\alpha & Q = 2.310\ MeV\ \text{(excited state, 94\%)} \end{cases} \tag{1}$$

$$ {}^{6}_{3}Li + {}^{1}_{0}n \rightarrow {}^{3}_{1}H + {}^{4}_{2}\alpha \qquad Q = 4.780\ MeV \tag{2}$$

$$ {}^{3}_{2}He + {}^{1}_{0}n \rightarrow {}^{3}_{1}H + {}^{1}_{1}p \qquad Q = 0.764\ MeV \tag{3}$$

The energy from the reaction is shared by the two reaction products, the *α*-particle or proton (*p*) and the recoil nucleus, according to conservation of momentum and energy.

The ^{6}Li(*n,α*) reaction is usually used in scintillators. One possibility is lithium iodide, which is chemically similar to sodium iodide. Due to the density of enriched ^{6}LiI(Eu) crystals, a 10 mm thick detector is almost 100% efficient for low energy neutrons ranging from thermal energies up to about 0.5 eV. Lithium is also incorporated as a component of scintillating glass matrices. Lithium glass scintillators are used in time-of-flight measurements due to their relatively fast time response of less than 100 ns. This type of detector, however, is more commonly used in the detection of neutrons with intermediate energies.

The ^{10}B(*n,α*) reaction is employed in BF_3 proportional tubes where the BF_3 gas is usually enriched in ^{10}B, and it has to be used at lower absolute pressures (between 0.5 and 1.0 atm) for performance and safety as a proportional gas counter. In a similar way, ^{3}He employing the ^{3}He (*n,p*) reaction is used as a conversion target and proportional gas, typically charged to between 2 and 10 atm for a ^{3}He proportional counter. Pressures greater than ~5 atm can yield diminishing returns in sensitivity[6], although this is very dependent on the moderator and geometry selected for the application.

The cross sections of the neutron capture reactions described in Equations 1, 2, and 3, respectively, tend to decrease rapidly with increasing neutron energy. Therefore the detectors mentioned earlier are very inefficient if used directly for neutrons of intermediate energies (also categorized as fast neutrons). But the slow neutron detectors can be surrounded by a hydrogen-containing material that moderates the neutrons down to energies where the detection efficiency is high. This moderation is achieved via elastic scattering, and neutrons can be slowed down most effectively by hydrogen nuclei, making polyethylene and paraffin the most commonly used moderators. The detection efficiency of a moderator-detector combination depends on the targeted neutron energy and the thickness of the moderator.[7,8]

Of the many types of neutron spectrometers that have been developed, the system known as the multi-sphere, or more commonly, the “Bonner” sphere (BS),[9] spectrometer has been built and used by more laboratories than any other, often with ^{3}He counters.[9, 10, 11] The detection system consists of a thermal neutron sensor used at the centre of a number of different diameter-moderating spheres. These are almost invariably made of polyethylene, and are usually made to exact inch, or half-inch diameters. The thermal sensor plus moderating sphere combination has sensitivity to neutrons over a broad energy range. However, the sensitivity for each sphere peaks at a particular neutron energy, depending on the sphere diameter. From the measured readings of a set of spheres, information can be derived about the spectrum of the neutron field in which the measurements were made. The derivation of this spectral information is not simple, and the validity of BS results has often been questioned[12] since BS systems work best when there is some a priori information about the spectra.[13] Among the designs that have evolved from the BS systems is the use of a single block of moderator containing either several extended, position-sensitive thermal neutron detectors[14], or a number of small thermal neutron detectors[15] mounted at different positions. The position of neutron detectors then replaces the sphere diameter as a variable in the response matrix, and all measurements are made simultaneously, without interruption for geometry changes.

One method of screening for certain isotopes involves characterization of neutron and γ-ray emission rates resulting from fissile sources, mostly spontaneous fission (s.f.) and alpha-neutron (α,n) reactions in the material. To yield an accurate profile of the neutron spectrum emanating from a fissile source, neutron self-multiplication (multiplicity) within a subcritical assembly must also be considered. Various configurations of moderating and attenuating materials can be placed around neutron sensitive detectors in order to isolate different parts of the neutron spectrum. In particular, the attribution of neutrons can be performed using 3-D numerical simulations to the radiation transport equation. To accomplish this, neutron transport must be simulated among a suite of moderators and detectors in a “transport optimized” detector array; this enables one to optimize neutron interactions to facilitate positive detection and complete characterization of the neutron spectrum. This raises the potential of not only detecting neutrons, but also of characterizing the SNM source that generated them spectrally, with the potential to discriminate between SNM metals and oxides, helpful for characterization of SNM material in scenarios of interest. In addition, gamma rays from fissile sources resulting from spontaneous fission and other interactions can also be accounted for to yield a complete mosaic of the radiation field from a fissile material source.[16, 17]

3. COMPUTATIONAL TRANSPORT METHODS

Accurate simulation of any particles transporting radiation in a system is directly accomplished via solution of the Boltzmann ("transport") equation,[18,19] which describes the behavior of neutral particles in terms of spatial, angular, and energy domains as they interact in a system; the steady-state form of the transport equation is given in Eq. 4 using standard notation:

$$\hat{\Omega}\cdot\nabla\psi(\vec{r},\hat{\Omega},E)+\sigma(\vec{r},E)\psi(\vec{r},\hat{\Omega},E)=\int_0^\infty\int_{4\pi} dE'\,d\Omega'\,\sigma_S\,(\vec{r},\hat{\Omega}'\cdot\hat{\Omega},E'\rightarrow E)\,\psi(\vec{r},\hat{\Omega}',E')$$
$$+\frac{\chi(\vec{r},E)}{k_0}\int_0^\infty dE'\,\nu\sigma_f\,(\vec{r},E')\,\phi(\vec{r},E')+Q(\vec{r},\hat{\Omega},E) \qquad (4)$$

The left side of Eq. 4 represents streaming and collision terms (loss), and the right side represents scattering and other sources (gain). Since it describes the flow of radiation in a three-dimensional (3-D) geometry with angular and energy dependence, this is one of the most challenging equations to solve in terms of complexity and model size, and rendering a deterministic computational solution for a large problem requires a robust parallel transport solver and a high performance computing system. Although typically less demanding to initiate, large scale Monte Carlo simulations also require significant computer resources and "smart" variance reduction methodologies to render credible, statistically based solutions. For the latest 3-D deterministic codes, PARTISN, PENTRAN, ATILLA, and DRAGON have emerged. PARTISN is a parallel code built from the THREEDANT code system, and has spatial parallel decomposition and time dependence capabilities. PENTRAN is a 3-D steady state code with parallel scalable memory and parallel decomposition among the angular, energy, and spatial variables, and an adaptive scheme for high accuracy spatial differencing. ATILLA is an unstructured (tetrahedral) mesh code that has been used for general applications in recent years. DRAGON was developed as a collision probability/method of characteristics code that has been widely used in CANDU reactor physics applications. [20,21,22,23] Among Monte Carlo codes, MCNP5/MCNPX are the most widely used codes for SNM detection assessments.[24]

3.1 Cross Section Library Considerations

Since computational transport methods are necessary to determine total leakage multiplication sources for specific source scenarios, adequate cross section libraries must be applied.[25,26,27] Monte Carlo users can readily access point-wise cross sections (as in MCNP5), although care needs to be taken to select appropriate thermal treatments [such as the 'S(α,β)' molecular scattering for moderators] and library temperatures. Also, deterministic cross section libraries must be evaluated carefully as to how they are applicable to a given problem. Resonance self shielding must be properly applied to produce multigroup cross section libraries that are adequate to describe applications under consideration. Reasonable agreement between Monte Carlo and deterministic results should be expected, and questions should be raised to explain differences of more than a few percent.

3.2 Total Source as a Function of Energy

To capture the spectral fidelity of SNM in radiation transport throughout an assembly, and to computationally determine the absolute scale of the detectable radiation leaking from SNM, the following procedure (for comparing S_N and Monte Carlo) may be applied: For direct comparison, energy bin tallies in Monte Carlo simulations must be equivalent to the deterministic (S_N) group structure. Compute via either deterministic or Monte Carlo radiation transport computations the energy group-dependent neutron leakage (signified by the group subscript g in Equation (5)) per induced fission reaction due to multiplication of the intrinsic source, where intrinsic sources include spontaneous fission (s.f.), alpha-neutron (α,n), etc. Then, add this to the leakage computed using the intrinsic source alone (also determined from 3-D transport computations)[4]:

$$M_{L,g_n} = \frac{\int_A \vec{J}\,|_{A \text{ induced}-n,g} \cdot d\vec{A}}{\sum_{g=1}^{G} \int_V Q_{\text{induced}-n,g}\, dV} \cdot \left(-\frac{Q_{o,n} V}{\rho} \right)_{\text{induced}-n} + \int_A \vec{J}\,|_{A \text{ intrinsic-}n,g} \cdot d\vec{A} \quad (5)$$

where:

$\int_V Q_{\text{induced}-n,g}\, dV$ = the energy group g induced fission neutron source from a criticality computation, where V constitutes the volume of SNM mass in the system;

$\int_A \vec{J}\,|_{A \text{ induced}-n,g} \cdot d\vec{A}$ = the energy group g induced fission neutron leakage at the SNM material surface, where A constitutes the surface area of the closed surface SNM mass in the system;

$\int_A \vec{J}\,|_{A \text{ intrinsic-}n,g} \cdot d\vec{A}$ = the energy group g intrinsic neutron leakage at the SNM material surface, where A constitutes the outer surface area of the SNM mass in the system;

$Q_{o,n}$ = intrinsic [s.f., (α,n) reactions] neutron volume source density (n/cm^{3}s^{-1});

$-\frac{Q_{o,n}}{\rho}$ = induced (multiplied) neutron source density (n/cm^{3}s^{-1});

$\rho = \left(\frac{k_{eff} - 1}{k_{eff}} \right)$ = the system reactivity based on fission multiplication.

Using Eq. (5), the total multiplied source leakage is determined in Eq. (6) from a sum of the total source from all energy groups:

$$M_{L,n} = \sum_{g=1}^{G} M_{L,g_n} \quad (6)$$

A similar set of equations describes the gamma rays (intrinsic and induced) leaking from the SNM system. Note that for an SNM mass in "free air", the net neutron or gamma currents crossing (leaking from) the outer SNM surface are approximately equal to the exiting (positive outward normal) positive partial current (see Eq. (7)), since almost no neutrons or gammas exiting from an assembly placed in air return into the multiplying system, e.g. ~1 in 10^4 neutrons, and ~0.5 in 10^4 gamma rays return for a 4 kg Pu (94% ^{239}Pu) alpha-metal sphere[4]:

$$\vec{J}_g\,|_A = (\vec{J}_g^{\,+}\,|_A - \vec{J}_g^{\,-}\,|_A) \approx \vec{J}_g^{\,+}\,|_A \tag{7}$$

Other objects and structures impacting the detection scenario will result in neutrons or gammas from higher energies streaming from the assembly to scatter in surrounding materials, lose energy, and subsequently appear in lower energies (higher energy *group* numbers), creating attributable re-entrant currents at the SNM surface. As mentioned, these must be evaluated on a case-by-case basis as a function of position (and time) for the model being studied, since multiplication can be affected.

3.3 Forward versus Adjoint Calculation Procedures

The steady state multi-group form of the transport equation operating on the forward group angular flux ψ_g is:

$$\begin{aligned}&\hat{\Omega}\cdot\nabla\psi_g(\vec{r},\hat{\Omega}) + \sigma_g(\vec{r})\psi_g(\vec{r},\hat{\Omega}) \\ &\qquad = \sum_{g'=1}^{G}\int_{4\pi} d\Omega'\,\sigma_{s\,g'\to g}(\vec{r},\hat{\Omega}'\cdot\hat{\Omega})\,\psi_g(\vec{r},\hat{\Omega}') + q_g(\vec{r},\hat{\Omega})\end{aligned} \tag{8}$$

Principally, scattering from all other groups *g'* into group *g*. is dominated by down-scattering from higher energies to lower energies. The adjoint transport operator H^+ can be derived using the adjoint identity for real valued functions, and the forward multi-group transport operator, where $\langle\ \rangle$ represents integration over all independent variables:

$$\left\langle \psi_g^+\, H\,\psi_g \right\rangle = \left\langle \psi_g\, H^+ \psi_g^+ \right\rangle \tag{9}$$

Using Eq. 8, it can be seen that the forward operator is

$$H = \hat{\Omega}\cdot\nabla + \sigma_g(\vec{r}) - \sum_{g'=1}^{G}\int_{4\pi} d\Omega'\,\sigma_{s\,g'\to g}(\vec{r},\hat{\Omega}'\cdot\hat{\Omega}) \tag{10}$$

The angular adjoint (importance) function is ψ_g^+. Applying the adjoint boundary condition that particles leaving a bounded system have an importance of zero in all groups (converse of the forward vacuum boundary condition) with the above equations, and requiring a continuous importance function mathematically leads to the multi-group adjoint transport operator:

$$H^{+} = -\hat{\Omega}\cdot\nabla + \sigma_g(\vec{r}) - \sum_{g'=1}^{G} \int_{4\pi} d\Omega' \sigma_{s\, g\rightarrow g'}(\vec{r}, \hat{\Omega}\cdot\hat{\Omega}') \tag{11}$$

Note the minus sign on the streaming term indicates that adjoint particles travel along a 'reversed' direction, where scattering progresses from group *g* *back* to other groups *g'* (those groups formerly contributing to group *g* in the forward equation). The adjoint function for our application is aliased to neutron importance with respect to an absorption in ^{3}He to yield an (*n,p*) reaction. If such a fixed forward neutron source/detector problem is proposed, the neutron flux must satisfy the transport equation:

$$H\psi_g = q_g \tag{12}$$

and the inhomogeneous adjoint equation should be satisfied with an adjoint source aliased to the group detector response cross section σ_{dg}:

$$H^{+}\psi_g^{+} = \sigma_{d\,\mathrm{g}} \tag{13}$$

Applying Equations 9, 12, and 13, and integrating over all variables results in the useful expression for detector response *R* for forward or adjoint computations:

$$R = \left\langle \psi_g \sigma_{d\,\mathrm{g}} \right\rangle = \left\langle \psi_g^{+} q_g \right\rangle \tag{14}$$

From Equation 14, it is clear that, detector response can be obtained by complete integration of the source distribution with the adjoint function—for any arbitrary source distribution. *R* can thereby be computed directly from the results of either of *several* forward transport computations for each neutron source, or one single adjoint transport computation with coupling to each source density[7].

3.4 Recent Transport Applications in SNM Monitoring and Nuclide Detection

With the combination of added emphasis in non-proliferation, homeland security needs, and large advances in computational capability, both deterministic and Monte Carlo radiation transport have seen wide application of late. Deterministic methods for neutron transport are again becoming more popular as the need for rapid computation while minimizing stochastic error in terms such as the fission source density are important. New advances in cross sections are enabling researchers to revisit computations of interest to improve predicted responses. For example, recent investigations by Kornreich with plutonium solutions have updated information on positive temperature coefficient feedback in various weak plutonium solutions, as verified with time dependent PARTISN computations.[28] Moreover, for neutron detection, Mattingly, et al developed new cross section libraries and a procedure for Feynman-Y (which tracks a departure from Poisson statistics) determinations in plutonium based multiplicity experiments simulated with deterministic PARTISN and a new Kynea3 library.[29]

In addition, deterministic methods for gamma transport are also important to current progress in the detection field. Mitchell and Mattingly developed a method to reconstruct gamma ray spectra simulations using one dimensional spherical ONEDANT computations that estimate currents from scattered radiation to compute shielded spectrum components. Using their method, an integral reconstruction of the gamma spectrum with shielding is accurately inferred.[30] Using a differential spectrum analysis approach with an intent to render enhanced photopeak identification, LaVigne, Sjoden, Baciak, and Detwiler developed a tool to post process gamma spectra to synthetically augment the spectral resolution to enhance photopeak extraction using precomputed Monte Carlo based detector response functions (DRFs) in a tool called ASEDRA. Complete pulse height simulated, aliased spectra attributed to photons are reconstructed extracted from the reference data spectra, revealing new spectral detail beneath, resulting in novel peak identification and dramatic enhancement of NaI(Tl) spectra.[31]

Kelley and Palmer, and Benz and Palmer recently implemented a deterministic particle balance pulse height reconstruction method for rapid deterministic spectral synthesis.[32,33] Additional improvement of DRFs using Monte Carlo transport was performed by Gardner's group to improve representation of sampling non-linearities in pulse height spectra in MCNP5.[34] DRFs were also recently constructed by Huh, Haghighat, and Baciak for BGO detectors using MCNP5 pulse height tallies.[35]

Monte Carlo has always been a staple for detection simulations, since with appropriate pointwise cross sections, one can exactly reproduce an experimental scenario with high detail on the computer, provided one can enable particles to penetrate the geometry adequately to yield reasonable statistics. Both the MCNP5 and MCNPX codes, developed and maintained by Los Alamos National Laboratory, are used extensively in non-proliferation and detection applications. Researchers have modified various versions of MCNP for their own purposes to enhance speed and accuracy. Wagner and Haghighat implemented the consistent adjoint driven importance sampling (CADIS) methodology using MCNP4A in the A^3MCNP code, which used an S_N adjoint solution from the TORT Code from Oak Ridge National Laboratory to appropriately bias the weight window technique.[36] Wagner extended this work with Peplow and Evans recently with the MAVRIC code sequence for the new SCALE6 coded system release. They implemented a new FW-CADIS approach that uses both forward and adjoint deterministic S_N computations simultaneously to augment convergence of near and far detectors in a Monte Carlo simulation.[37]

Pozzi, Padovani, and Marseguerra developed the MCNP-PoliMi version from the standard MCNP code version to simulate each neutron–nucleus interaction and photon production are correlated, so that fully consistent neutron and photon fission multiplicities have been implemented. This enables detailed information (energy, particles, locations, etc) about each collision interaction to be specifically tracked. Such information is needed in correlating neutron and gamma outputs in multiplicity measurements.[38] Subsequently, Henkel and Mihalczo recently applied MCNP-PoliMi to model prompt-neutron time behavior after interrogating an HEU metal casting using time-tagged Cf-252 neutrons.[39] In addition, Dimitrov, et al, recently introduced a GUI-driven wrapper for MCNPX that contains material and geometry libraries so as to enable simulation of smuggling scenarios to assess threats, actively running MCNPX in the background to determine responses.[40]

Significant work is ongoing to merge the MCNP5 and MCNPX codes to be released as MCNP6. In particular, researchers collaborating between University of Illinois and Los Alamos have improved consistency of charged particle stopping powers, particularly for protons and alpha particles in various media, so that accuracies of charged particle transport are improved by as much as 25%.[41]

Accounting for 3-D fuel depletion and transmutation as it relates to validating safeguards and security protocols for stored spent fuel is an important question, particularly with increasing spent fuel/on-site storage used fuel inventories. In fact, this places new emphasis on extremely accurate whole core fuel cycle modeling, which drives the need for parallel computing and more efficient and accurate assessment tools. 3-D deterministic codes that can handle this issue include DRAGON, PENTRAN/PENBURN, and ATILLA. For Monte Carlo problems in burnup, SCALE6/KENO and MCNPX incorporate burnup/depletion sequences.

Finally, breakthroughs in new sources for laboratory testing and validation have also been recently developed. Researchers at the FINDS Institute in Florida have developed a source driven by WGPu-Be generated alpha-neutrons that shifts the harder alpha-neutron source spectrum to a close alias to a fission source; this source was named "JEZZEBALL". This means that a reliable fission source surrogate, previously only available via a fixed source of Cf-252 and a half life of 2.65 years, is now available for use based on the 24,110 year half life decay of Pu-239. Using this source, Ghita, et al, have carried out a detailed moderator study, and have identified a four "energy band" approach to isolate portions of SNM neutron spectra using ^{3}He. As validation of computational results is required for field trials, more practical yet realistic sources similar to JEZZEBALL will be needed in non-proliferation, safeguards, and homeland security applications.[42]

4. CONCLUSIONS

The use of transport theory methods, particularly 3-D Deterministic radiation transport, as a tool in assessing scenarios involving Special Nuclear Materials (SNM) detection for homeland security and non-proliferation purposes has increased significantly in recent years. This is primarily due to the need to optimize detectors and understand radiation signatures to overcome the many challenges associated with SNM detection, as well as understand the complex nature of the radiation spectra involving neutron and gamma transport in detection scenarios. Augmenting this is the more-ready access and availability of high performance computing. Overall, the valuable information obtained from deterministic and Monte Carlo transport methods, and how those methods have been recently applied, were summarized.

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